

Two-Photon Interferometry for High-Resolution Imaging

Dmitry Strekalov and Jonathan Dowling

JPL, MS 300-123,
4800 Oak Grove Drive,
Pasadena, CA 91109.

Dmitry.V.Strekalov@jpl.nasa.gov

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Abstract

We discuss advantages of using non-classical states of light for two aspects of optical imaging: creating of miniature images on photosensitive substrates, which constitutes the foundation for optical lithography, and conversely, imaging of micro objects. In both cases, the classical resolution limit given by the Rayleigh criterion is approximately a half of the optical wavelength. It has been shown, however, that by using multi-photon quantum states of the light field, and multi-photon sensitive material or detector, this limit can be surpassed. In the present work, we give a rigorous quantum mechanical treatment of this problem, address some particularly widespread misconceptions and discuss the requirements arising on the way of turning the research on quantum imaging into a practical technology.

The idea that the limits of classical optical imaging can be overcome by using multi-photon processes is fairly well known. For example, Marlan Scully discusses in his book [1] a two-photon microscope scheme that would beat the width limitation of the standard diffraction pattern by a factor of $\sqrt{2}$ by making a $\text{sinc}^4(kx)$ diffraction pattern, which comes as a product of two classical diffraction patterns, instead of the $\text{sinc}^2(kx)$. Such narrowing of a diffraction pattern can be observed by using a detector sensitive to intensity-square, instead of just intensity. In other words, one needs a two-photon process to observe the $\sqrt{2}$ -narrowing of diffraction pattern. Moreover, using detectors based on a higher-order multi-photon process that are sensitive exclusively to an even higher order of intensity, one could see even narrower diffraction patterns.

This approach would not work so well for holographic imaging used in lithography. In this technique, the desired image is composed of interference fringes of different spatial frequency, so the resolution is given by the highest spatial frequency. The spatial frequency is equal to the inverse of the fringe period, which cannot be shorter than one half of the optical wavelength. It is easy to see that this period is the same for any power of intensity, e.g. a $\sin^4(kx)$ fringe has the same period as a $\sin^2(kx)$ fringe.

Different approaches have been suggested to obtain a shorter-period interference fringe of intensity-square. It has been proposed, for example, to use frequency modulation to blur

the longer spatial component of a $\sin^4(kx)$ fringe, [2]. Using quantum sources of light has been also proposed [3] and demonstrated with electronic detection of coincidences [4].

Consider the setup that has been proposed for quantum interferometric lithography [3], see Fig.1. This is a modification of a well-known two-photon interference experiment [5,6], in which the single-photon detectors are removed, and the output beams are directed at a two-photon sensitive substrate (e.g., one covered with a lithographical photoresist) instead.

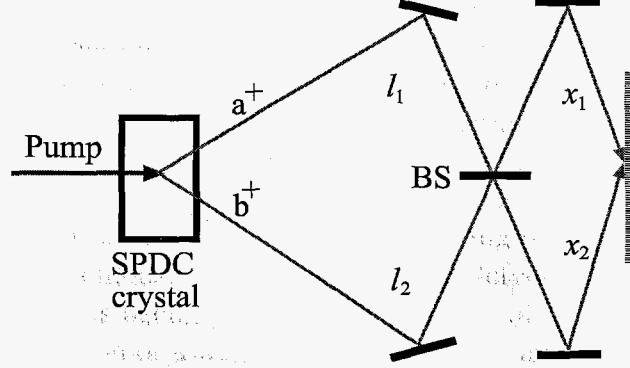


Figure 1: Two-photon interferometer with photosensitive substrate.

Following the standard theoretical treatment for two-photon interferometers, we write the two-photon amplitude-square as

$$|A|^2 \equiv \langle \Psi | \hat{E}^{(-)} \hat{E}^{(-)} \hat{E}^{(+)} \hat{E}^{(+)} | \Psi \rangle = |\langle 0 | \hat{E}^{(+)} \hat{E}^{(+)} | \Psi \rangle|^2, \quad (1)$$

where the fields depend on the propagation paths, and the state $|\Psi\rangle$ is the frequency-entangled output state of a Spontaneous Parametric Down Converter (SPDC):

$$|\Psi\rangle = \int d\nu h(\nu) \hat{a}^\dagger(\nu) \hat{b}^\dagger(-\nu) |0\rangle. \quad (2)$$

In (2), creation operators \hat{a}^\dagger and \hat{b}^\dagger refer to channels labeled l_1 and l_2 , respectively, in Fig.1; ν is the frequency-detuning from the central frequency ω_0 , the later being equal to one half of the pump frequency ω_p . The spectral function $h(\nu)$ gives the phase matching width and accounts for inexact momentum conservation due to the finite length of the crystal L :

$$h(\nu) = \frac{1 - e^{-iL\Delta_z(\nu)}}{iL\Delta_z(\nu)}. \quad (3)$$

Derivation and analysis of expressions (2) and (3) are given in a number of publications on SPDC. In particular, in [7,8], it is shown that for collinear degenerate type-I SPDC

$$\Delta_z(\nu) = -D'\nu^2, \quad D' = \frac{d}{d\omega} \frac{1}{v} \Big|_{\omega_0}, \quad (4)$$

and for collinear degenerate type-II SPDC

$$\Delta_z(\nu) = D\nu, \quad D = \frac{1}{v_o} - \frac{1}{v_e}, \quad (5)$$

where v denotes the group velocity of the signal and idler photons. In case of orthogonal polarizations (type-II), the group velocity v has indices o and e for "ordinary" and "extraordinary" polarization components.

The two-photon amplitude (1) can describe coincidence detection rate, as well as the two-photon absorption rate, as a function of pathlengths $l_{1,2}$ and $x_{1,2}$. In the coincidence detection case, the fields in (1) are evaluated at two distinct locations of two detectors, while in the two-photon absorption case they are evaluated at the same, although arbitrary, point on the photosensitive substrate. A geometrical size of the "point" in this context may be equal to the size of photo emulsion grain, or of the photoresist molecule. Let us assume that this size is much smaller than the interference structure we are expecting to see. Then we can safely speak of a zero-size point. As a further simplification, we will consider a one-dimensional problem with exactly counterpropagating beams. This geometry is obviously not practical, since no light energy is delivered to the surface, and we study this case just as an illustration allowing us to simplify the treatment.

As a next step, we need to represent the fields in (1) in terms of the same operators that describe the two-photon wavefunction (2). For perfectly monochromatic plane waves with a wave vector $k = \omega_0/c$, the connection is given by propagating the operators through the interferometer:

$$E^{(+)} = \hat{a}e^{ikl_1} \left(\frac{1}{\sqrt{2}}e^{ikx_2} + \frac{i}{\sqrt{2}}e^{ikx_1} \right) + \hat{b}e^{ikl_2} \left(\frac{1}{\sqrt{2}}e^{ikx_1} + \frac{i}{\sqrt{2}}e^{ikx_2} \right). \quad (6)$$

In equation (6), we put the proportionality constant between the field operator and the annihilation operator equal to unity. Also, we assume that the fields in the arms l_1 and l_2 have the same polarization. It is easy to see that otherwise there will be no two-photon interference fringes on the photosensitive substrate.

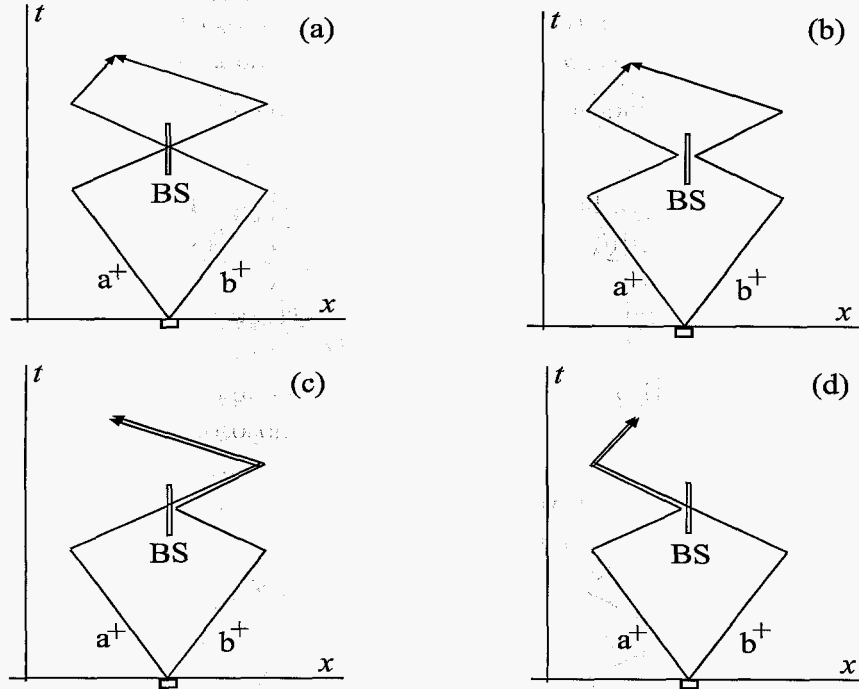


Figure 2: Different two-photon paths contributing to the amplitude (1): (a) both photons are transmitted; (b) both reflected; (c) transmitted - reflected; (d) reflected - transmitted.

The plane wave approximation implies that in the wave function (2), $h(\nu)$ should be replaced by $\delta(\nu)$. Then substituting (6) and (2) into (1) it is easy to notice that the terms

with \hat{a}^2 and \hat{b}^2 drop out, which is consistent with only one photon present in each channel. The other four terms can be represented by four paths shown in Fig.2. These paths correspond to both photons being transmitted by the beamsplitter (a), both reflected by it (b), one transmitted, the other reflected (c), and vice versa (d). If we have a 50-50 beamsplitter, all four amplitudes are equally weighted. Notice that, in the usual coincidence-detection treatment of two-photon interference, the amplitudes corresponding to paths (c) and (d) are discarded simply because they do not result in a pair of coincident detections. Therefore one cannot directly apply the results well known for a two-detectors experiment to our system, and then argue that "detectors are placed at the same point", since at that point the amplitudes (c) and (d) are already discarded. Let us now show that these are the amplitudes that give rise to two-photon interference.

In the following, we will consider a more realistic case of wavepackets rather than plane waves. The fields will be allowed to have a finite frequency bandwidth, described by a real and even function $f(\nu)$, around the central frequency ω_0 :

$$E = \int d\nu f(\nu) \left\{ \hat{a}(\nu) e^{ik(\nu)l_1} \left(e^{ik(\nu)x_2} + i e^{ik(\nu)x_1} \right) + \hat{b}(\nu) e^{ik(\nu)l_2} \left(e^{ik(\nu)x_1} + i e^{ik(\nu)x_2} \right) \right\}. \quad (7)$$

Then the two-photon amplitude (1) takes on the following form:

$$A = \int d\nu d\nu_1 d\nu_2 h(\nu) f(\nu_1) f(\nu_2) \left\{ e^{ik(\nu_1)l_1} e^{ik(\nu_2)l_2} \left(e^{ik(\nu_1)x_2} + i e^{ik(\nu_1)x_1} \right) \left(e^{ik(\nu_2)x_1} + i e^{ik(\nu_2)x_2} \right) \right\} \langle 0 | \hat{a}(\nu_1) \hat{b}(\nu_2) \hat{a}^\dagger(\nu) \hat{b}^\dagger(-\nu) | 0 \rangle. \quad (8)$$

The inner product in (8) is equal to $\delta(\nu_1 - \nu) \delta(\nu_2 + \nu)$ which reduces (8) to a single integral. To handle it, we expand $k(\nu) = k_0 + \nu/c$, where $k_0 \equiv k(\omega_0)$. This allows us to arrive at,

$$A = e^{ik_0(l+x)} [u(\Delta l + \Delta x) - u(\Delta l - \Delta x) - 2 \sin(2k_0 \Delta x) u(\Delta l)], \quad (9)$$

where $u(z)$ is given by a Fourier transformation of a combined spectral density, and therefore has a meaning of a correlation function:

$$u(z) \equiv \int d\nu h(\nu) f^2(\nu) e^{i\frac{\nu}{c}z}. \quad (10)$$

In (10), $x \equiv x_2 + x_1$, $\Delta x \equiv x_2 - x_1$, $l \equiv l_2 + l_1$ and $\Delta l \equiv l_2 - l_1$ have been introduced.

Analyzing the symmetry properties of $h(\nu)$, we find that in both cases of type-I (4) and type-II (5) SPDC $u(z)$ is always a real and even function:

$$u(z) = u(-z) = u^*(z) = u^*(-z). \quad (11)$$

Therefore the first two terms in (9) cancel each other when $\Delta l = 0$. Taking the absolute square of the remaining term, we get

$$|A|^2 = 4u^2(0) \sin^2(2k_0 \Delta x). \quad (12)$$

We see from (12) that the two-photon absorption amplitude is a harmonic function of coordinate Δx measured along the photosensitive substrate, that has a spatial frequency

$4k_0$, which is twice the spatial frequency of the "usual", second-order interference fringes. The two-photon interference fringes (12) appear to have a perfect contrast for all Δx . This is a consequence of a plane wave approximation *for the pump*. If one considers the pump with a finite bandwidth, the exponential pre-factor in (9) will no longer be just a phase factor, but will turn into an envelope, equivalent to the pump envelope. Therefore the two-photon interference fringes (12) will have a coherence length equal to the pump coherence length, which may be quite long and reach meters for CW lasers.

It is very important that the two-photon coherence length does not depend on the fields bandwidth given by $f(\nu)$, nor on the phase matching width given by $h(\nu)$. Why this happens is obvious from the physical consequences of the condition $\Delta l = 0$. It has been shown [5, 6], that in this case the two-photon amplitudes represented in Fig.2 by diagrams (a) and (b) exactly cancel each other, and the photon pair always goes to one channel (either x_1 or x_2), realizing diagrams (c) or (d). In other words [3], the beamsplitter produces an entangled $|2\rangle_{x_1}|0\rangle_{x_2} - |0\rangle_{x_1}|2\rangle_{x_2}$ state, which picks up the spatial phase at the same rate as the pump photon would. It also dephases at the same slow rate as the pump photon does due to its finite bandwidth, which results in the two-photon coherence length of the SPDC light being equal to the pump (single-photon) coherence length.

Now let us consider the linear interference in our apparatus. This study is important, since the modulations of intensity will directly affect the result (12) for two-photon absorption rate. For example, there will be no two-photon absorption in the nodes of the single-photon interference fringe.

The expression for intensity is

$$I = \langle \Psi | E^{(-)} E^{(+)} | \Psi \rangle, \quad (13)$$

where the state $|\Psi\rangle$ is given by (2) and the field is given by (7). Setting $l_1 = l_2$ and treating this expression the same way we have treated the forth-order field momenta, we arrive to

$$I = 1 - \cos(k_0 \Delta x) \int d\nu |h(\nu)|^2 f^2(\nu) \sin\left(\frac{\nu}{c} \Delta x\right). \quad (14)$$

Notice that the integrand in (14) is an odd function, and hence the whole integral is zero, and the expression (14) equals unity. This means that in our apparatus there will be no intensity modulations due to the second-order interference, regardless of individual coherence length of the signal and idler photons. This at first appears surprising, since one might expect to see at least a few interference fringes at the "white light interference" condition $x_1 = x_2$. However, taking into account that both inputs of the beamsplitter are used, we realize that we actually have two sets of interference fringes exactly out of phase with each other, and hence the total intensity is unmodulated.

Two more issues associated with two-photon quantum imaging need to be addressed to make it a practically useful technology. One is availability of two-photon sensitive photoreists and detectors, and the other has to do with the fact that using SPDC as a two-photon source, one first loses a factor of two in spatial resolution by down converting the pump frequency (and hence doubling the wavelength), and then re-gains this factor by using two-photon processes. Therefore in terms of spatial resolution, quantum imaging technique has no advantage over using classical imaging at the pump wavelength. The counter argument [9] is that it is not always possible to use the UV light. For example, it may be incompatible with imaging biological or other light sensitive objects. Another example is

3D lithography [9]: creating 3D structures with single-photon exposure of photolithographical materials is very difficult since they strongly absorb UV light which limits the depth of penetration. Two-photon exposures solve this problem. However, much of the value would be added to quantum imaging technology if one could prepare two-photon states without doubling the wavelength. One way to achieve it is to use a Hyper Parametric Scattering (HPS) instead of SPDC.

HPS is a nonlinear optical process occurring via the cubical optical nonlinearity $\chi^{(3)}$, in which *two* pump photons recombine into an entangled photon pair. This process is similar to four wave mixing in the same sense as SPDC is similar to Parametric Amplification: four wave mixing and PA assume non-vacuum input into the signal and/or the idler modes. HPS is distinct from the SPDC where *a single* pump photon produces an entangled pair. This distinction is the most evident from comparing the phase matching conditions for SPDC:

$$\vec{k}_p = \vec{k}_s + \vec{k}_i, \quad \omega_p = \omega_s + \omega_i \quad (15)$$

with those for HPS:

$$2\vec{k}_p = \vec{k}_s + \vec{k}_i, \quad 2\omega_p = \omega_s + \omega_i \quad (16)$$

which is illustrated graphically in Fig.3. An important thing to notice in Fig.3 is that the average wavelength of the photons produced in HPS is the same as that of the pump, while in the case of SPDC it doubles.

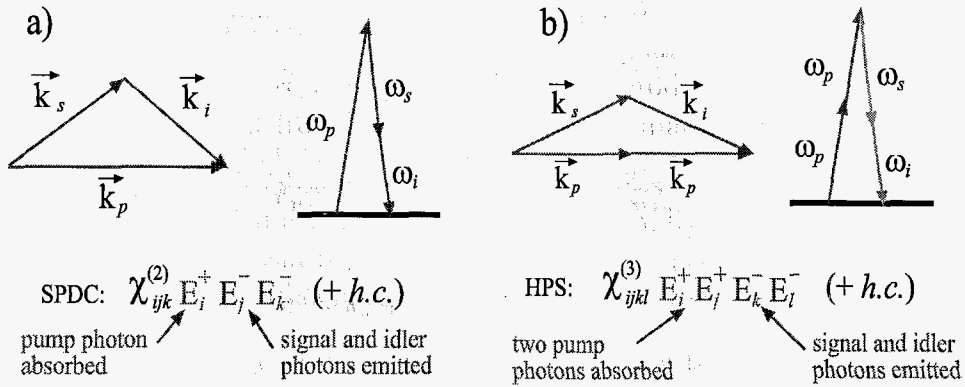


Figure 3: The phase matching (momentum and energy conservation) diagrams for SPDC (a) and HPS (b).

For the first time, HPS was observed over 30 year ago [10]. At that time, it did not attract the due attention as a source of EPR-states because of a very low efficiency of the $\chi^{(3)}$ processes compared to $\chi^{(2)}$ processes. A typical value for $\chi^{(2)}$ is 10^{-8} [CGS units of electric field] $^{-1}$, while for $\chi^{(3)}$ it is 10^{-15} [CGS units of field] $^{-2}$. Fortunately, HPS output power is quadratic with respect to the pump intensity, while in case of SPDC it is only linear, so comparing efficiencies of the two processes, one compares the squares of $E_p \chi^{(2)}$ and $\chi^{(3)}$. Modern powerful femtosecond lasers, that were not yet available in the early days of the HPS discovery, dramatically changed the situation in favor of HPS.

Another argument in favor of HPS is that unlike SPDC, this process does not require any particular symmetry of the media, and can be observed not only in crystals but also in glass fibers [11], which promises to increase the interaction length to meters, or beyond. Furthermore, it has been shown [12] that about a four orders of magnitude improvement of

the signal can be achieved by cascading two $\chi^{(2)}$ processes to emulate a $\chi^{(3)}$ HPS process. Large amount of other research has been done on $\chi^{(3)}$ processes, and particularly on four wave mixing, e.g. [11, 13–15], and we plan to rely on these results in our new research program directed at creating a robust source of entangled photon pairs or two-photon states without down converting the light frequency.

The second practical issue we have mentioned above is availability of two-photon sensitive photoresists. Considering very low power of two-photon sources, high two-photon sensitivity of the photoresists is required. Unfortunately, high single-photon UV sensitivity of many commercially available photoresists does not guaranty that they would be suitable two-photon sensitive materials. Synthesizing of such a material appears to be a difficult task, although a large volume of research has been done in this area, e.g. [9, 16, 17], motivated by the growing recognition of the two-photon imaging technology importance.

We also have carried out a preliminary quick search for two-photon sensitive lithographic materials. Relying on analogy with atomic systems, we expect that a suitable two-photon material would have an intermediate level corresponding to the single photon energy, so that the single-photon detuning that inversely factors in the two-photon absorption cross section be small, and the two-photon absorption rate be peaked. It is furthermore required that the molecular transition corresponding to the intermediate absorption level does not result in the photochemical reaction initiating the phototresist (otherwise the resist would be one-photon sensitive); that the intermediate level or band is normally depopulated and very short-lived (otherwise the resist would be one-photon sensitive via cascaded processes); and that both transitions have the right selection rules.

We have taken absorption spectra of various commercially available photoresists. The results are shown in Fig.4. One of our samples, the Novalac 5740, has shown a local absorption maximum which is centered at about 520 nm and is clearly separated from the strong transition in the UV part of the spectra, associated with the photochemical reaction initiating the photoresist. We span an approximately 15 μm -thick sample of this photoresist on a gold plated substrate and exposed the sample to different doses of the Argon Ion laser light, whose wavelength (514.5 nm) was close to the center of the absorption peak of interest. We found the threshold dose of about 2 kJ/cm², assuming 100% radiation reflection off the mirror substrate and operating at the intensity level of 5 W/cm². Repeating the experiment at 25 W/cm², we obtained the same results with five times shorter exposition time, which suggests that the exposure process is linear in intensity and hence is a single-photon one. Notice that the threshold we found at 514.5 nm is roughly five orders of magnitude higher than for a regular UV exposure.

Next, we repeated the expositions for another Argon Ion laser line with the 457.9 nm wavelength, which is off the intermediate absorption peak but is closer to the UV absorption transition. We found that at this wavelength the threshold dose was definitely lower than 0.4 kJ/cm². This suggests that the high-threshold phototinitiation observed at 514.5 nm, as well as at 457.9 nm, is not related to the intermediate absorption peak, but rather is due to a far off-resonant absorption on the wing of the UV absorbing transition. Therefore, the selected material may satisfy the above-outlined requirements for a two-photon optimized photoresist, and it would be interesting to try exposing it with a two-photon source. We plan on carrying out such experiment in the nearest future.

In conclusion, we have carried out a rigorous analysis that confirmed the results [3]. In addition, our analysis have shown that the desired two-photon interference fringe will have

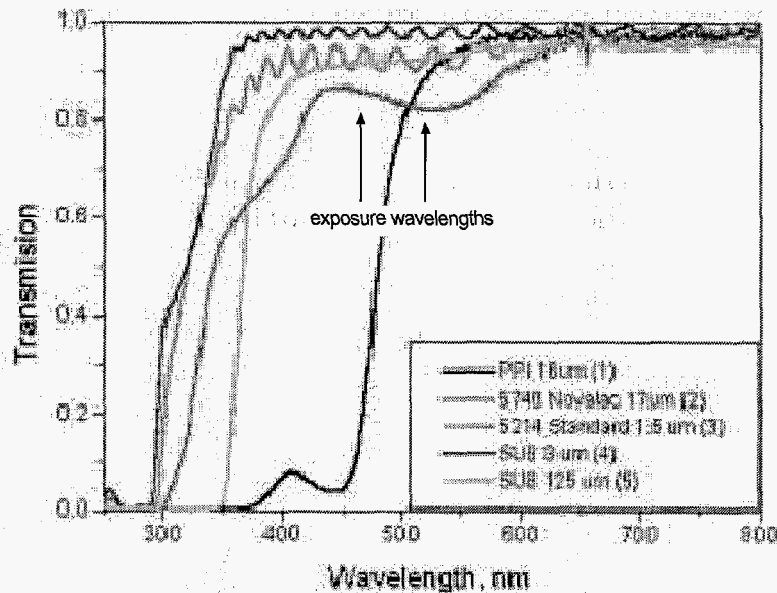


Figure 4: Absorption spectra for different photoresists. The sample of the choice shows an absorption maximum centered at about 520 nm. Arrows mark the wavelengths the sample was exposed at: 457.9 nm and 514.5 nm.

a very long coherence length, equal to that of the pump, and that the second-order (single-photon) interference fringes will be entirely absent. The questions related to alternative sources of two-photon states and to the choice of two-photon sensitive photolithographical materials have been discussed. Although bringing the research in this area to the level of practical technology is a challenging task, it is at the same time is an interesting and potentially rewarding one.

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